

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

Applicant: Younes Bouizi,

Examiner: James CORNO.

Serial No.: 10/587,854

Group Art Unit: 1793

Filed: October 14, 2008

For : CATALYST IN THE FORM OF GRAINS COMPRISING AN ACIDIC POROUS CORE SURROUNDED BY A UNIFORM EXTERNAL LAYER.

**DECLARATION UNDER 37 C.F.R. §1.132**

Assistant Commissioner for Patents

Washington, D.C. 20231

Sir :

I, Slavik KASZTELAN, being duly warned, declare and say as follows:

THAT, I am a French citizen holding the titles of Engineer delivered by "Ecole des Hautes Etudes Industrielles de Lille" in 1982, of "Docteur ès Sciences" delivered by "Université de Lille" in 1984, of "Doctor accredited to supervise research" delivered by "Université de Lille" in 1991, residing at 69006 Lyon, France, 97 quai Charles de Gaulle.

THAT, I have been engaged on research by "Institut Français du Pétrole" in their Kinetics and Catalysis Department since 1988, where I have been continuously and actively in charge of researches in the fields of hydrocracking, hydroisomerization, dewaxing and hydrogenation of aromatic compounds. I was since September 2002 to April 2006 the manager of the Division "Catalysis and Separation". I am since May 1<sup>st</sup>, 2006, the Assistant Manager of the whole Refining and Petrochemical Division.

I declare further :

THAT, I am familiar with the content of U.S. Patent Application Serial No 10/587,854.

THAT, I know the teaching of US Patent N0. 6.872.865.

THAT, I declare below the technical reasons supporting the patentability of the claimed invention.

US 6.872.865 discloses a catalyst for the disproportionation of aromatic hydrocarbons. Said catalyst is a layered crystalline composite of a single zeolitic isotype comprising a core which comprises a zeolitic aluminosilicate and a mantle comprising a crystalline metallosilicate, preferably boralite, containing a framework metal capable of forming stable +3, preferably B (col 2 lines 23-31). US 6.872.865 insists on an essential feature : the composite is of "a common crystal structure or zeolitic isotype" (col 3 lines 22-23, col 4 lines 29-30 and line 64) and the phases of the composite are crystallographically indistinct (col 3 lines 28-29). The catalyst disclosed in US 6.872.865 is prepared according to a process involving the crystal growth of the mantle phase in the presence of the core (col 3 lines 19-22). The core is "in integral lattice association or epitaxial alignment" with the mantle (col 3 lines 13-17). This means that the crystallographic orientations of the crystals of the mantle are aligned with the crystals of the core due to the isotypical character between the core and the mantle and the grown layer (epitaxial growth). It is known that the faces of one crystal do not all present the same crystallographic orientations and that the growth rate is higher for some faces than others. Then, in the process disclosed in US 6.872.865, the growth rate of the mantle depends on the crystalline faces of the core and leads to a layer thickness varying according to the different crystallographic orientations of the core crystals. Hence, this direct growth of crystalline metallosilicalite (mantle) on a zeolitic aluminosilicalite (core) is not a suitable method for obtaining a uniform thickness of the mantle covering the core.

The present invention claims a catalyst in the form of grain, each grain being formed by an acidic crystallized microporous zeolitic core covered by at least one uniform external layer made of a crystallized microporous zeolitic solid. An essential feature of the claimed catalyst is that the zeolite of the core differs from the zeolite of the external layer by the structural type so that the crystallographic structure of the core is different from the one of the external layer. Therefore the claimed catalyst is different from the catalyst as disclosed in US 6.872.865. The preparation process of the claimed catalyst includes a multi-stage procedure with the following major steps : a) preparation of crystals or crystal agglomerates to form the crystallized microporous zeolitic core b) modification of the core to impart at least partial

acidity to said core c) uniform deposition and adhesion of nanosized nuclei based on the crystallized microporous zeolitic solid of the external layer over the surface of the core, d) growth of said nuclei on the core. Said preparation process as described in the specification as filed allows to obtain a composite catalyst in which 1) the core and the external layer are fully crystallographically independent while US 6.872.865 teaches the opposite, 2) the crystallographic orientation of the external layer is not controlled by the core (as it is the case in US 6.872.865 where the catalyst is prepared by epitaxial growth) but by the nanocrystals based on the crystallized microporous zeolitic solid of the external layer fixed on the core. The growth of the nuclei in step d) depends on the crystalline faces of nuclei but, contrary to the teaching of US 6.872.865, the crystalline faces of said nuclei are disconnected from the crystalline structure and the crystallographic orientation of the faces of the crystals of the core. Consequently, such a preparation process involving a specific nanosized nuclei adhesion procedure with fixation of nanocrystals from which the external layer grows leads to a composite catalyst in which an integral lattice association or epitaxial alignment of the core with the external layer cannot be observed. Due to uniform deposition and adhesion of the nuclei on the outer surface of the core, performed in said step c), using notably the reverse electrostatic charge polymer, the layer is uniform whatever the crystallographic face of the crystals of the core.

For all these reasons, the claimed catalyst is new in view of US 6.872.865 and is not at all suggested in US 6.872.865 which teaches the skilled worker away from preparing a composite catalyst in which the core and the external layer does not have the same crystallographic structure.

The other cited references cannot render the claimed invention obvious, even in combination with US 6.872.865 : US 5.082.814 discloses a composite catalyst in which the outer layer is not made of a zeolite and Twaiq's reference teaches a composite material in which the outer layer is made of a mesoporous material which cannot be a zeolite since it is known that zeolite are microporous materials (Molecular Sieves, Second edition 1998, page 22 (chapter 1.6.3. microporous materials)).

The undersigned declares further that all statements made herein of this own knowledge are true and that all statements made on information or belief are believed to be true; and further

that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 Title 18 of United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Solaize, December 18, 2009.

A handwritten signature in cursive script, appearing to read "Slavik Kasztelan".

**Slavik KASZTELAN**